Practitioner's Docket No. U 013829-3

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00140

PATENT TRADEMARK OFFICE

CHAPTER II

TRANSMITTAL LETTER TO THE UNITED STATES ELECTED OFFICE (EO/US)

(ENTRY INTO U.S. NATIONAL PHASE UNDER CHAPTER II)

PCT/ES00/00260	19 JULY 2000	21 JULY 1999
INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED

MATERIAL USEFUL AS ELECTRIC CONDUCTOR AND COMPRISING POLYACETYLENE OR SUBSTITUTED POLYACETYLENES INCORPORATED IN A MESOPOROUS SUPPORT, PRODUCTION PROCESS, AND CONDUCTORS MADE OF SAID MATERIAL

TITLE OF INVENTION

1. Mercedes ALVARO RODRIGUEZ; 2. Hermenegildo GARCIA GOMEZ; 3. Andrew GILBERT; 4. David CARDIN

APPLICANT(S)

Box PCT
Assistant Commissioner for Patents
Washington D.C. 20231
ATTENTION: EO/US

CERTIFICATION UNDER 37 C.F.R. 1.10*
*(Express Mail label number is **mandatory**.)*
(Express Mail certification is optional.)

I hereby certify that this correspondence and the documents referred to as attached therein are being deposited with the United States Postal Service on this date JANUARY 15, 2002, in an envelope as "Express Mail Post Office to Addressee," Mailing Label Number EV011020160US, addressed to the: Assistant Commissioner for Patents, Washington, D.C. 20231.

JENNIFER RASHKIN
(type or print name of person mailing paper)

Signature of person mailing paper

WARNING: *Certificate of mailing (first class) or facsimile transmission procedures of 37 C F.R 1 8 cannot be used to obtain a date of mailing or transmission for this correspondence.*

***WARNING:** *Each paper or fee filed by "Express Mail" **must** have the number of the "Express Mail" mailing label placed thereon prior to mailing. 37 C F R 1.10(b).*
*"Since the filing of correspondence under § 1 10 without the Express Mail mailing label thereon is an oversight that can be avoided by the exercise of reasonable care, requests for waiver of this requirement will **not** be granted on petition." Notice of Oct. 24, 1996, 60 Fed. Reg. 56,439, at 56,442.*

NOTE: The completion of those filing requirements that can be made at a time later than 30 months from the priority date results from the Commissioner exercising his judgment under the authority granted under 35 USC 371(d). The filing receipt will show the actual date of receipt of the last item completing the entry into the national phase. See 37 C.F.R. §1.491 which states: "An international application enters the national state when the applicant has filed the documents and fees required by 35 USC 371(c) within the periods set forth in § 1.494 and § 1 495."

WARNING: *Where the items are those which can be submitted to complete the entry of the international application into the national phase are subsequent to 30 months from the priority date the application is still considered to be in the international state and if mailing procedures are utilized to obtain a date the express mail procedure of 37 C.F.R. §1.10 must be used (since international application papers are not covered by an ordinary certificate of mailing - See 37 C F.R. §1 8.*

NOTE. Documents and fees must be clearly identified as a submission to enter the national state under 35 USC 371 otherwise the submission will be considered as being made under 35 USC 111 37 C F.R § 1.494(f)

1. Applicant herewith submits to the United States Elected Office (EO/US) the following items under 35 U.S.C. 371:

   a. [X] This express request to immediately begin national examination procedures (35 U.S.C. 371(f)).

   b. [X] The U.S. National Fee (35 U.S.C. 371(c)(1)) and other fees (37 C.F.R. § 1.492) as indicated below:

2.Fees

CLAIMS FEE	(1) FOR	(2) NUMBER FILED	(3) NUMBER EXTRA	(4) RATE	(5) CALCULATIONS
	TOTAL CLAIMS*	41- 20 =	21	x $ 18.00 =	NOT PAID
	INDEPENDENT CLAIMS*	2- 3 =		x $ 84.00 =	
	MULTIPLE DEPENDENT CLAIM(S) (if applicable) + $280.00				
BASIC FEE**	[ ] U.S. PTO WAS INTERNATIONAL PRELIMINARY EXAMINATION AUTHORITY Where an International preliminary examination fee as set forth in § 1.482 has been paid on the international application to the U.S. PTO [ ] and the international preliminary examination report states that the criteria of novelty, inventive step (non-obviousness) and industrial activity, as defined in PCT Article 33(2) to (4) have been satisfied for all the claims presented in the application entering the national stage (37 CFR 1.492(a)(4)) ..... $100.00 [ ] and the above requirements are not met (37 CFR 1.492(a)(1)) ..... $710.00 [X] U.S. PTO WAS NOT INTERNATIONAL PRELIMINARY EXAMINATION AUTHORITY Where no international preliminary examination fee as set forth in § 1.482 has been paid to the U.S. PTO, and payment of an international search fee as set forth in § 1.445(a)(2) to the U.S. PTO: [ ] has been paid (37 CFR 1.492(a)(2)) ..... $740.00 [X] has not been paid (37 CFR 1.492(a)(3)) ..... $1,040.00 [ ] where a search report on the international application has been prepared by the European Patent Office or the Japanese Patent Office (37 CFR 1.492(a)(5)) $890.00				
	Total of above Calculations				=$1040.00
SMALL ENTITY	Reduction by ½ for filing by small entity, if applicable. Statement may also be filed. (note 37 CFR 1.9, 1.27, 1.28)				-
	Subtotal				$1040.00
	Total National Fee				$1040.00
	Fee for recording the enclosed assignment document $40.00 (37 CFR 1.21(h)). (See Item 13 below). See attached "ASSIGNMENT COVER SHEET".				
TOTAL	Total Fees enclosed				$1040.00

*May include Preliminary Amendment (see page 8) reducing the number of claims.

i. [X] A check in the amount of $1040.00 to cover the above fees is enclosed.

ii. [ ] Please charge Account No. _____ in the amount of $ _____. A duplicate copy of this sheet is enclosed.

**WARNING: *"To avoid abandonment of the application the applicant shall furnish to the United States Patent and Trademark Office not later than the expiration of 30 months from the priority date * * * (2) the basic national fee (see § 1.492(a)). The 30-month time limit may not be extended " 37 C F R § 1 495(b)*

WARNING: *If the translation of the international application and/or the oath or declaration have not been submitted by the applicant within thirty (30) months from the priority date, such requirements may be met within a time period set by the Office 37 C F R § 1 495(b)(2) The payment of the surcharge set forth in § 1.492(e) is required as a condition for accepting the oath or declaration later than thirty (30) months after the priority date. The payment of the processing fee set forth in § 1.492(f) is required for acceptance of an English translation later than thirty (30) months after the priority date Failure to comply with these requirements will result in abandonment of the application. The provisions of § 1.136 apply to the period which is set. Notice of Jan. 3, 1993, 1147 O.G. 29 to 40.*

[ ] Applicant hereby asserts status as a small entity under 37 C.F.R. § 1.27.
[ ] A Statement or Written Assertion is attached.

NOTE: *37 C.F.R. § 1.27(c) deals with the assertion of small entity status, whether by a written specific declaration thereof or by payment as a small entity of the basic filing fee or the fee for the entry into the national phase as states:*

"(c) Assertion of small entity status Any party (person, small business concern or nonprofit organization) should make a determination, pursuant to paragraph (f) of this section, of entitlement to be accorded small entity status based on the definitions set forth in paragraph (a) of this section, and must, in order to establish small entity status for the purpose of paying small entity fees, actually make an assertion of entitlement to small entity status, in the manner set forth in paragraph (c)(1) or (c)(3) of this section, in the application or patent in which such small entity fees are to be paid.

(1) Assertion by writing. Small entity status may be established by a written assertion of entitlement to small entity status. A written assertion must:

(i) Be clearly identifiable,

(ii) Be signed (see paragraph (c)(2) of this section); and

(iii) Convey the concept of entitlement to small entity status, such as by stating that applicant is a small entity, or that small entity status is entitled to be asserted for the application or patent. While no specific words or wording are required to assert small entity status, the intent to assert small entity status must be clearly indicated in order to comply with the assertion requirement

(2) Parties who can sign and file the written assertion. The written assertion can be signed by:

(i) One of the parties identified in §§ 1.33(b) (e.g , an attorney or agent registered with the Office), §§ 3.73(b) of this chapter notwithstanding, who can also file the written assertion;

(ii) At least one of the individuals identified as an inventor (even though a §§ 1.63 executed oath or declaration has not been submitted), notwithstanding §§ 1.33(b)(4), who can also file the written assertion pursuant to the exception under §§ 1.33(b) of this part; or

(iii) An assignee of an undivided part interest, notwithstanding §§ 1.33(b)(3) and 3./73(b) of this chapter, but the partial assignee cannot file the assertion without resort to a party identified under §§ 1.33(b) of this part

(3) Assertion by payment of the small entity basic filing or basic national fee The payment, by any party, of the exact amount of one of the small entity basic filing fees set forth in §§ 1.16(a), (f), (g), (h), or (k), or one of the small entity basic national fees set forth in §§ 1.492(a)(1), (a)(2), (a)(3), (a)(4) or (a)(5), will be treated as a written assertion of entitlement to small entity status even if the type of basic filing or basic national fee is inadvertently selected in error.

(i) If the Office accords small entity status based on payment of a small entity basic filing or basic national fee under paragraph (c)(3) of this section that is not applicable to that application, any balance of the small entity fee that is applicable to that application will be due along with the appropriate surcharge set forth in §§ 1.16(e) or §§ 1.16(l).

(ii) The payment of any small entity fee other than those set forth in paragraph (c)(3) of this section (whether in the exact fee amount or not) will not be treated as a written assertion of entitlement to small entity status and will not be sufficient to establish small entity status in an application or a patent "

3. [X] A copy of the International application as filed (35 U.S.C. 371(c)(2)):

NOTE. Section 1.495 (b) was amended to require that the basic national fee and a copy of the international application must be filed with the Office by 30 months from the priority date to avoid abandonment "The International Bureau normally provides the copy of the international application to the Office in accordance with PCT Article 20. At the same time, the International Bureau notifies applicant of the communication to the Office. In accordance with PCT Rule 47.1, that notice shall be accepted by all designated offices as conclusive evidence that the communication has duly taken place. Thus, if the applicant desires to enter the national stage, the applicant normally need only check to be sure the notice from the International Bureau has been received and then pay the basic national fee by 30 months from the priority date." Notice of Jan. 7, 1993, 1147 O.G. 29 to 40, at 35-36. See item 14c below.

a. [ ] is transmitted herewith.
b. [ ] is not required, as the application was filed with the United States Receiving Office.
c. [X] has been transmitted
   i. [X] by the International Bureau.
   Date of mailing of the application (from form PCT/IB/308): _____.
   ii. [ ] by applicant on _____.
   Date

4. [X] A translation of the International application into the English language (35 U.S.C. 371(c)(2)):
a. [X] is transmitted herewith.
b. [ ] is not required as the application was filed in English.
c. [ ] was previously transmitted by applicant on _____.
   Date
d. [ ] will follow.

5. [X] Amendments to the claims of the International application under PCT Article 19 (35 U.S.C. 371(c)(3)):

NOTE. The Notice of January 7, 1993 points out that 37 C F R § 1.495(a) was amended to clarify the existing and continuing practice that PCT Article 19 amendments must be submitted by 30 months from the priority date and this deadline may not be extended. The Notice further advises that, "The failure to do so will not result in loss of the subject matter of the PCT Article 19 amendments. Applicant may submit that subject matter in a preliminary amendment filed under section 1.121. In many cases, filing an amendment under section 1 121 is preferable since grammatical or idiomatic errors may be corrected." 1147 O.G. 29-40, at 36.

a. [ ] are transmitted herewith.
b. [ ] have been transmitted
   i. [ ] by the International Bureau.
   Date of mailing of the amendment (from form PCT/IB/308): _____.
   ii. [ ] by applicant on _____.
   Date
c. [X] have not been transmitted as
   i. [X] applicant chose not to make amendments under PCT Article 19.
   Date of mailing of Search Report (from form PCT/ISA/210):
   12 SEPTEMBER 2000 _____.
   ii. [ ] the time limit for the submission of amendments has not yet expired. The amendments or a statement that amendments have not been made will be transmitted before the expiration of the time limit under PCT Rule 46.1.

6. [X] A translation of the amendments to the claims under PCT Article 19 (38 U.S.C. 371(c)(3)):
a. [ ] is transmitted herewith.
b. [ ] is not required as the amendments were made in the English language.
c. [X] has not been transmitted for reasons indicated at point 5(c) above.

7. [X] A copy of the international examination report (PCT/IPEA/409)
[X] is transmitted herewith.
[ ] is not required as the application was filed with the United States Receiving Office.

8. [ ] Annex(es) to the international preliminary examination report
a. [ ] is/are transmitted herewith.
b. [ ] is/are not required as the application was filed with the United States Receiving Office.

9. [ ] A translation of the annexes to the international preliminary examination report
a. [ ] is transmitted herewith.
b. [ ] is not required as the annexes are in the English language.

10. [X] An oath or declaration of the inventor (35 U.S.C. 371(c)(4)) complying with 35 U.S.C. 115
    a. [ ] was previously submitted by applicant on _____. Date
    b. [ ] is submitted herewith, and such oath or declaration
        i. [ ] is attached to the application.
        ii. [ ] identifies the application and any amendments under PCT Article 19 that were transmitted as stated in points 3(b) or 3(c) and 5(b); and states that they were reviewed by the inventor as required by 37 C.F.R. 1.70.
    c. [X] will follow.

Other document(s) or information included:

11. [X] An International Search Report (PCT/ISA/210) or Declaration under PCT Article 17(2)(a):
    a. [X] is transmitted herewith.
    b. [ ] has been transmitted by the International Bureau. Date of mailing (from form PCT/IB/308): _____.
    c. [ ] is not required, as the application was searched by the United States International Searching Authority.
    d. [ ] will be transmitted promptly upon request.
    e. [ ] has been submitted by applicant on _____. Date

12. [X] An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98:
    a. [ ] is transmitted herewith. Also transmitted herewith is/are:
        [ ] Form PTO-1449 (PTO/SB/08A and 08B).
        [ ] Copies of citations listed.
    b. [X] will be transmitted within THREE MONTHS of the date of submission of requirements under 35 U.S.C. 371(c).
    c. [ ] was previously submitted by applicant on _____. Date

13. [ ] An assignment document is transmitted herewith for recording.

A separate [ ] "COVER SHEET FOR ASSIGNMENT (DOCUMENT) ACCOMPANYING NEW PATENT APPLICATION" or [ ] FORM PTO 1595 is also attached.

_____
_____
_____

14. [X] Additional documents:
    a. [X] Copy of request (PCT/RO/101)
    b. [X] International Publication No. WO 01/05840
        i. [X] Specification, claims and drawing
        ii. [ ] Front page only
    c. [X] Preliminary amendment (37 C.F.R. § 1.121)
    d. [X] Other

    FORM PCT/IPEA/401

15. [X] The above checked items are being transmitted
    a. [X] before 30 months from any claimed priority date.
    b. [ ] after 30 months.

16. [ ] Certain requirements under 35 U.S.C. 371 were previously submitted by the applicant on _____, namely:

AUTHORIZATION TO CHARGE ADDITIONAL FEES

WARNING: *Accurately count claims, especially multiple dependent claims, to avoid unexpected high charges if extra claims are authorized.*

NOTE: "A written request may be submitted in an application that is an authorization to treat any concurrent or future reply, requiring a petition for an extension of time under this paragraph for its timely submission, as incorporating a petition for extension of time for the appropriate length of time An authorization to charge all required fees, fees under § 1 17, or all required extension of time fees will be treated as a constructive petition for an extension of time in any concurrent or future reply requiring a petition for an extension of time under this paragraph for its timely submission Submission of the fee set forth in § 1 17(a) will also be treated as a constructive petition for an extension of time in any concurrent reply requiring a petition for an extension of time under this paragraph for its timely submission." 37 C.F.R. § 1.136(a)(3).

NOTE: "Amounts of twenty-five dollars or less will not be returned unless specifically requested within a reasonable time, nor will the payer be notified of such amounts, amounts over twenty-five dollars may be returned by check or, if requested, by credit to a deposit account." 37 C F R § 1.26(a)

[X] The Commissioner is hereby authorized to charge the following additional fees that may be required by this paper and during the entire pendency of this application to Account No. 12-0425 .

[X] 37 C.F.R. 1.492(a)(1), (2), (3), and (4) (filing fees)

WARNING: *Because failure to pay the national fee within 30 months without extension (37 C.F.R § 1.495(b)(2)) results in abandonment of the application, it would be best to always check the above box.*

[ ] 37 C.F.R. 1.492(b), (c) and (d) (presentation of extra claims)

NOTE: Because additional fees for excess or multiple dependent claims not paid on filing or on later presentation must only

be paid or these claims cancelled by amendment prior to the expiration of the time period set for response by the PTO in any notice of fee deficiency (37 C.F.R § 1.492(d)), it might be best not to authorize the PTO to charge additional claim fees, except possible when dealing with amendments after final action.

[X] 37 C.F.R. 1.17 (application processing fees)
[X] 37 C.F.R. 1.17(a)(1)-(5)(extension fees pursuant to § 1.136(a).
[X] 37 C.F.R. 1.18 (issue fee at or before mailing of Notice of Allowance, pursuant to 37 C.F.R. 1.311(b))

NOTE: *Where an authorization to charge the issue fee to a deposit account has been filed before the mailing of a Notice of Allowance, the issue fee will be automatically charged to the deposit account at the time of mailing the notice of allowance 37 C F.R. § 1.311(b)*

NOTE *37 C F.R 1 28(b) requires "Notification of any change in loss of entitlement to small entity status must be filed in the application . . . prior to paying, or at the time of paying . . . issue fee." From the wording of 37 C.F.R. § 1.28(b). (a) notification of change of status must be made even if the fee is paid as "other than a small entity" and (b) no notification is required if the change is to another small entity.*

[ ] 37 C.F.R. § 1.492(e) and (f) (surcharge fees for filing the declaration and/or filing an English translation of an International Application later than 30 months after the priority date).

SIGNATURE OF PRACTITIONER

Reg. No.: 25,858

William R. Evans
(type or print name of practitioner)

Tel. No.: (212) 708-1930

P.O. Address

Customer No.: 00140

c/o Ladas & Parry
26 West 61st Street
New York, N.Y. 10023

10031057.020902 #5

Practitioner's Docket No. U 013829-3 ***PATENT***

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

[X] In re application of: Mercedes ALVARO RODRIGUEZ, et al.
International Application No.: PCT/ES00/00260 Group No.:
International Filing Date: 19 July 2000 Examiner:
For: MATERIAL USEFUL AS ELECTRIC CONDUCTOR AND COMPRISING POLYACETYLENE OR SUBSTITUTED POLYACETYLENES INCORPORATED IN A MESOPOROUS SUPPORT, PRODUCTION PROCESS, AND CONDUCTORS MADE OF SAID MATERIAL

[ ] *Patent No.: Issue Date:

**NOTE.* *Insert name(s) of inventor(s) and title also for patent Where statement is with respect to a maintenance fee payment, also insert application number and filing date, and add Box M Fee to address*

STATEMENT CLAIMING SMALL ENTITY STATUS (37 CFR 1.9(c-f) and 1.27(b-d))

With respect to the invention described in
[ ] the specification filed herewith.
[x] application no. PCT/ES00/00260, filed 19 July 2000.
[ ] patent no. _____ issued _____.

I. IDENTIFICATION AND RIGHTS AS A SMALL ENTITY

I hereby state that I am

(complete either (a), (b), (c) or (d) below)

(a) Independent Inventor
[ ] a below named independent inventor, and that I qualify as an independent inventor, as defined in 37 CFR 1.9(c), for purposes of paying reduced fees under Sections 41(a) and (b) of Title 35, United States Code, to the Patent and Trademark Office.

(b) Noninventor Supporting a Claim by Another
[ ] making this statement to support a claim by

_____

for a small entity status for purposes of paying reduced fees under Sections 41(a) and (b) of Title 35, United States Code. I hereby state that I would qualify as an independent inventor as defined in 37 CFR 1.9(c) for purposes of paying reduced fees under Sections 41(a) and (b) of Title 35, United States Code, if I had made the above identified invention.

(c) Small Business Concern
[ ] the owner of the small business concern identified below:
check one → [ ] an official of the small business concern empowered to act on behalf of the concern identified below:

EXPRESS MAIL LABEL NO.: EV 011023081 US

Name of Concern_____
Address of Concern_____
_____and
that the above identified small business concern qualifies as a small business concern, as defined in 13 CFR 121.3-18, and reproduced in 37 CFR 1.9(d), for purposes of paying reduced fees under Sections 41(a) and (b) of Title 35, United States Code, in that the number of employees of the concern, including those of its affiliates, does not exceed 500 persons. For purposes of this statement, (1) the number of employees of the business concern is the average over the previous fiscal year of the concern of the persons employed on a full-time, part-time or temporary basis during each of the pay periods of the fiscal year, and (2) concerns are affiliates of each other when either, directly or indirectly, one concern controls or has the power to control the other, or a third party or parties controls or has the power to control both.

(d) Non-Profit Organization

[X] an official empowered to act on behalf of the nonprofit organization identified below:

Name of Organization UNIVERSIDAD POLITECNICA DE VALENCIA
Address of Organization Camino de Vera, E-46022 Valencia, SPAIN
_____

TYPE OF ORGANIZATION

[X] University or Other Institution of Higher Education

[ ] Tax Exempt Under Internal Revenue Service Code (26 USC 501(a) and 501(c) (3))

[ ] Nonprofit Scientific or Educational Under Statute of State of the United States of America
(Name of State_____)
(Citation of Statute_____)

[ ] Would Qualify as Tax Exempt Under Internal Revenue Service Code (26 USC 501(a) and 501(c) (3)), if Located in the United States of America

[ ] Would Qualify as Nonprofit Scientific or Educational Under Statute of State of the United States of America, if Located in the United States of America
(Name of State_____)
(Citation of Statute_____)

and that the nonprofit organization identified above qualifies as a nonprofit organization, as defined in 37 CFR 1.9(e), for purposes of paying reduced fees under Sections 41(a) and (b) of Title 35, United States Code.

II. OWNERSHIP OF INVENTION BY DECLARANT

I hereby state that rights under contract or law remain with and/or have been conveyed to the above identified

[ ] person (item (a) or (b) above)	[ ] concern (item (c) above)	[X] organization (item (d) above)

EXCEPT, that if the rights held are not exclusive, each individual, concern or organization having rights to the invention is listed below* and no rights to the invention are held (1) by any person who could not be classified as an independent inventor under 37 CFR 1.9(c) if that person had made the invention, (2) any concern which would not qualify as a small business concern under 37 CFR 1.9(d) or (3) a nonprofit organization under 37 CFR 1.9(e).

[X] no such person, concern, or organization
[ ] person, concerns or organizations listed below*

**NOTE: Separate statements are required from each named person, concern or organization having rights to the invention as to their status as small entities. (37 CFR 1 27)*

Full Name _____
Address _____
[ ] INDIVIDUAL [ ] SMALL BUSINESS CONCERN [ ] NONPROFIT ORGANIZATION

Full Name _____
Address _____
[ ] INDIVIDUAL [ ] SMALL BUSINESS CONCERN [ ] NONPROFIT ORGANIZATION

III. ACKNOWLEDGEMENT OF DUTY TO NOTIFY PTO OF STATUS CHANGE

I acknowledge the duty to file, in this application or patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28(b))

IV. DECLARATION

(check the following item, if desired)

NOTE: The following verification statement need not be made in accordance with the rules published on October 10, 1997, 62 Fed. Reg. 52131, effective December 1, 1997

NOTE. "The presentation to the Office (whether by signing, filing, submitting, or later advocating) of any paper by a party, whether a practitioner or non-practitioner, constitutes a certification under § 10 18(b) of this chapter. Violations of § 10 18(b)(2) of this chapter by a party, whether a practitioner or non-practitioner, may result in the imposition of sanctions under § 10.18(c) of this chapter. Any practitioner violating § 10 18(b) may also be subject to disciplinary action. See §§ 10.18(d) and 10.23(c)(15)." 37 CFR 1 4(d)(2).

[ ] I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this verified statement is directed.

V. SIGNATURES

(complete only (e) or (f) below)

(e)
NOTE: All inventors must sign the statement.

_____
Name of Inventor

_____ Date: _____
Signature of Inventor

_____
Name of Inventor

_____ Date: _____
Signature of Inventor

_____
Name of Inventor

_____ Date: _____
Signature of Inventor

(add lines for any additional inventors who must sign)

or

(f)
NOTE: The title of the person signing on behalf of a concern or nonprofit organization should be specified

Name of Person Signing (X) JUSTO NIETO NIETO

Title of Person (X) Rector
(if signing on behalf of a concern or non-profit organization)

Address of Person Signing Camino de Vera, E-46022 Valencia, SPAIN

SIGNATURE (X) _____ DATE (X) 3 July 2002

#2

Practitioner's Docket No. U 013829-3

CHAPTER II

IN THE UNITED STATES ELECTED OFFICE (EO/US)

PCT/ES00/00260	19 JULY 2000	21 JULY 1999
INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED

MATERIAL USEFUL AS ELECTRIC CONDUCTOR AND COMPRISING POLYACETYLENE OR SUBSTITUTED POLYACETYLENES INCORPORATED IN A MESOPOROUS SUPPORT, PRODUCTION PROCESS, AND CONDUCTORS MADE OF SAID MATERIAL
TITLE OF INVENTION

1. Mercedes ALVARO RODRIGUEZ; 2. Hermenegildo GARCIA GOMEZ; 3. Andrew GILBERT; 4. David CARDIN
APPLICANT(S)

Box PCT
Assistant Commissioner for Patents
Washington, D.C. 20231
ATTENTION: EO/US

PRELIMINARY AMENDMENT

Please amend the above identified application as follows:

IN THE CLAIMS :

Please amend claims 4-9, 11, 13, 15-16, 19-21, 27-28, 31-35, 37 and 39 as follows:

4. (Amended) A material according to claim 1, characterised in that it comprises an aluminosilicate host which incorporates a polymer comprised by polyacetylene substituted with phenyl groups.

CERTIFICATE UNDER 37 1.10

I hereby certify that this paper is being deposited with the United States Postal Service on this date JANUARY 15, 2002 in an envelope as "EXPRESS MAIL POST OFFICE TO ADDRESSEE" Mailing Label Number EV011020160US addressed to the: Assistant Commissioner for Patents, Washington, D.C. 20231

JENNIFER RASHKIN
(Type or print name of person mailing paper)

(Signature of person mailing paper)

NOTE: Each paper or fee referred to as enclosed herein has the number of the "EXPRESS MAIL" mailing label place thereon prior to mailing 37 CFR 1.16(b).

5. (Amended) A material according to claim 1, characterised in that the host is an hexagonal-phase aluminosilicate of the MCM-41 type.

6. (Amended) A material according to claim 1, characterised in tha tthe host is a cubic-phase uminosilicate of the MCM-48 type.

7. (Amended) A material according to claim 1, characterised in that the aluminosilicate has been modified by ion-exchange with transition metal ions.

8. (Amended) A material according to claim 1, characterised in that the aluminosilicate has been modified by impregnation by means of the introduction of transition metal ions.

9. (Amended) A material according to claim 7, characterised in that the metal ions are Ni^{2+} ions.

11. (Amended) A material according to claim 7, characterised in that the metal ions are Fe^{3+} ions.

13. (Amended) A material according to claim 7, characterised in that the metal ions are Cu^{2+} ions.

15. (Amended) A material according to claim 1, characterised in that the polymer exhibits a positive charge consisting in a p-doping.

16. (Amended) A material according to claim 1, characterised in that the polymer exhibits a negative charge consisting in an n-doping.

19. (Amended) A process according to claim 16, characterised in that the aluminosilicate is modified by ion-exchange with transition metal ions in an aqueous solution prior to the polymerization phase.

20. (Amended) A process according to claim 16, characterised in that the aluminosilicate is modified by impregnation by means of the introduction of transition metal ions.

21. (Amended) A process according to claim 19, characterised in that the metal ions are selected from Ni^{2+}, Fe^{3+} and Cu^{2+} ions.

27. (Amended) A process according to claim 25, characterised in that the doping is carried out electrochemically.

28. (Amended) A process according to claim 25, characterised in that the doping is carried out chemically.

31. (Amended) An electrical conductor characterised in that it comprises a material according to claim 1.

32. (Amended) An electrical conductor characterised in that it comprises a material prepared according to the process according to claim 17.

33. (Amended) A conductor according to claim 31, characterised in that it is an electrode used in batteries, which comprises said material.

34. (Amended) A conductor according to claim 31, characterised in that it is a conductive surface for displays, which comprises said material.

35. (Amended) A conductor according to claim 31, characterised in that it is a conductive surface for luminous devices, which comprises said material.

37. (Amended) A conductor according to claim 33, characterised in that the electrode comprises

an inert, vitrified and polished electrode

a coating of the inert electrode at least composed by a mixture of said material, powdered graphite and paraffin as an agglutinant.

39. (Amended) A conductor according to claim 34, characterised in that the conducting surface is a layer obtained by the deposition on a host of a suspension of said material on an organic solvent with an aqueous solution of polyvinyl alcohol.

Remarks

The above amendatory action is taken solely for the purpose of avoiding claim fees that would otherwise accrue due to the presence of multiple dependent claims.

Respectfully submitted,

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4. (Amended) A material according to claim[s] 1,[2 or 3,] characterised in that it comprises an aluminosilicate host which incorporates a polymer comprised by polyacetylene substituted with phenyl groups.

5. (Amended) A material according to [any of] claim[s] 1 [to 4], characterised in that the host is an hexagonal-phase aluminosilicate of the MCM-41 type.

6. (Amended) A material according to [any of] claim[s] 1 [to 4], characterised in tha tthe host is a cubic-phase uminosilicate of the MCM-48 type.

7. (Amended) A material according to [any of] claim[s] 1 [to 6], characterised in that the aluminosilicate has been modified by ion-exchange with transition metal ions.

8. (Amended) A material according to [any of] claim[s] 1 [to 6], characterised in that the aluminosilicate has been modified by impregnation by means of the introduction of transition metal ions.

9. (Amended) A material according to claim[s] 7 [or 8], characterised in that the metal ions are Ni^{2+} ions.

11. (Amended) A material according to claim[s] 7 [or 8] , characterised in that the metal ions are Fe^{3+} ions.

13. (Amended) A material according to claim[s] 7 [or 8], characterised in that the metal ions are Cu^{2+} ions.

15. (Amended) A material according to claim[s] 1 [to 4] , characterised in that the polymer exhibits a positive charge consisting in a p-doping.

16. (Amended) A material according to claim[s] 1 [to 4], characterised in that the polymer exhibits a negative charge consisting in an n-doping.

19. (Amended) A process according to [any of] claim[s] 16 [to 18], characterised in that the aluminosilicate is modified by ion-exchange with transition metal ions in an aqueous solution prior to the polymerization phase.

20. (Amended) A process according to [any of] claim[s] 16 [to 18], characterised in that the aluminosilicate is modified by impregnation by means of the introduction of transition metal ions.

21. (Amended) A process according to claim 19 [or 20], characterised in that the metal ions are selected from Ni^{2+}, Fe^{3+} and Cu^{2+} ions.

27. (Amended) A process according to claim[s] 25 [or 26], characterised in that the doping is carried out electrochemically.

28. (Amended) A process according to claim[s] 25 [or 26], characterised in that the doping is carried out chemically.

31. (Amended) An electrical conductor characterised in that it comprises a material according to [any of] claim[s] 1[-16].

32. (Amended) An electrical conductor characterised in that it comprises a material prepared according to the process according to [any of] claim[s] 17[-30].

33. (Amended) A conductor according to claim[s] 31 [or 32], characterised in that it is an electrode used in batteries, which comprises said material.

34. (Amended) A conductor according to claim[s] 31 [or 32], characterised in that it is a conductive surface for displays, which comprises said material.

35. (Amended) A conductor according to claim[s] 31 [or 32], characterised in that it is a conductive surface for luminous devices, which comprises said material.

37. (Amended) A conductor according to claim[s] 33 [or 36], characterised in that the electrode comprises

an inert, vitrified and polished electrode

a coating of the inert electrode at least composed by a mixture of said material, powdered graphite and paraffin as an agglutinant.

39. (Amended) A conductor according to claim[s] 34 [or 35], characterised in that the conducting surface is a layer obtained by the deposition on a host of a suspension of said material on an organic solvent with an aqueous solution of polyvinyl alcohol.

TITLE OF THE INVENTION

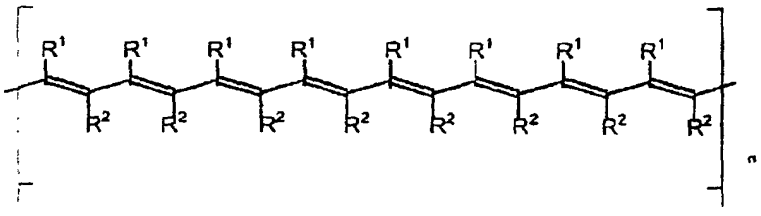
A material useful as an electrical conductor which comprises polyacetylene or substituted polyacetylenes incorporated in a mesoporous host, process for obtaining the material and conductors comprising the material.

TECHNICAL FIELD OF THE INVENTION

The present invention refers to electrical conducting organic polymers, particularly to conducting materials composed by an inorganic host and specifically to conducting materials in which the inorganic host incorporates an electrically conducting polymer.

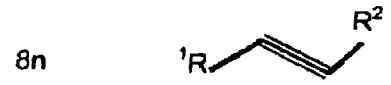
STATE OF THE ART PRIOR TO THE INVENTION

There are a series of organic polymers which under certain oxidation states exhibit a high electrical conductivity and which, for this reason, are called conducting organic polymers (Novák, Müller et al. "Electrochemically Active Polymers for Rechargeable Batteries" (1997), Chem. Rev. 97:207-281). Although in such polymers the monomeric unit which repeats itself throughout the polymer can be very varied, structurally they have in common the presence of a linear substructure of conjugated C=C double bonds. Among these conducting organic polymers, the most simple one, and for which a highest electrical conductivity has been described, includes polyacetylene and substituted polyacetylenes which present the structural formula:



and which is obtained from the polymerisation of alkine

monomers with the structural formula:



Where R^1 and R^2 are H or a substituent such as alkyl groups of less than four carbons (Me, Eth, Pr, i-Pr, Bu), phenyl groups with or without substituents at the para position (with groups such as Me, Eth, MeO, HO, etc), cyano group and methoxycarbonyl.

The properties of the basic polyacetylene chain can be controlled by the presence of electron-donor or electron-acceptor substituents. To this respect, the synthesis of substituted polyacetylenes from suitable polyacetylenes by using wolframium and molybdenum transition metals has been described (Masuda and Higashimura: "Polyacetylenes with Substituents: Their Synthesis and Properties" (1987), Adv. Polym. Sci., 81: 121-165).

The existence of extended conjugation throughout the conducting polymer is not enough to provide polyacetylene with a high electrical conductivity since in neutral state these materials behave as insulators. It is therefore necessary to proceed to the introduction of electrical charges (= doping) into the polymer, be it positive charges (= p type doping) or negative (= n type doping). Conductivity manifests itself after a specific charge threshold.

Although from a practical point of view, conducting organic polymers could be applied as substitutes of metallic conductors in printed circuits and as electrodes, their application in the industrial practice has been essentially limited due to two reasons: i) the difficulties in the synthesis of polyacetylene and its derivatives, and ii) the great instability of polyacetylenes, particularly after being doped, which gives rise to their oxidative degradation in the presence of oxygen, which results in the destruction of the polyacetylene chain by reaction with molecular oxygen. This

results in that polyacetylenes and their derivatives must be manipulated in inert gas atmosphere or under a vacuum, which considerably complicates the manufacture and handling of conducting elements which employ these materials.

Materials which combine conducting organic polymers with mineral hosts have been described (Cox and Stucky: "Polymerization of Methylacetylene in Hydrogen Zeolites" (1991), J. Phys. Chem., 95:710-720; Pereira, Kokotailo et al.: "Acetylene Polymerization in a H-ZSM-5 Zeolite" (1991), J. Mater. Chem., 3(8):867-872; Lewis, Millar et al.: "Trans-Polyacetylene on Sodium and Caesium Mordenites: A Resonance Raman Spectroscopic Study" (1993), Chem. Mat., 5:1509-1517; Millar, Lewis et al: "Raman-Spectroscopic Study of the Formation of Polyacetylene within Zeolite Channels" (1993), J. Mater. Chem., 3:867-872) obtained by the preparation of polyacetylene and polypropyne in the channels of microporous zeolites of medium pore size, that is, ZSM-5 Zeolite with elliptical channels of 0.54 x 0.56 nm, and of greater pore size, i.e. mordenite with 0.74 nm mean diameter channels.

The direct polymerization of pyrrole and thiophene to respectively give polypyrrole and polythiophene in the interior of the MCM-41 mesoporous aluminosilicate has also been described (Wu and Bein: "Conducting Carbon Wires in Ordered Nanometer-Sized Channels" (1994), Science, 266:705-709: Wu and Hein: "Conducting Polymer Wires in Mesopore Hosts" (1994), Stud. Surf. Sci. Catal. 84:2269-2276; Wu and Bein "Polyaniline Wires in Oxidant-Containing Mesoporous Channel Hosts" (1994), Chem. Mater., 6(8): 1109-1112; McCann, Millar et al.: "Formation of Polypyrrole and Polythiophene within Cu^{2+} Mordenite and H^{+} Mordenite Hosts Studied by EPR and UV-VIS Spectroscopy" (1995), J. Chem. Soc. Faraday Trans., 91(23): 4321-4328).

The previously mentioned materials do not permit to obtain sufficiently high contents of conducting organic polymers in the inorganic hosts so as to find utility in industrial applications. Therefore, the inner volume available for polymerization in zeolites and mordenites is

insufficient to manage to house amounts of conducting polymer greater than 10%, which in addition is achieved without obtaining polymers with sufficient purity. Additionally, the reduced size of the pores of the microporous zeolite hosts does not allow the hosts to house organic polymers substituted with voluminous groups, such as for example phenyl (or other substituted phenyls of formula $R\text{-}C_6\text{-}H_4$ where R can be an alkyl group of less than four carbons, as well as a methoxy or hydroxy group).

On the other hand, polypyrrole and polythiophene polymers are structurally different from polyacetylene and its derivatives.

OBJECTS OF THE INVENTION

The present invention has the object of providing materials useful as electrical conductors which comprise a host incorporating a conducting organic polymer, polyacetylene or polyacetylene derivatives, which protects the polymer from oxidation by oxygen in the environment, which are easy to manufacture and which are susceptible of containing the conducting polymer in a proportion sufficient to be able to take advantage if their conductive properties, and which permit the doping of the conducting polymer in a simple and effective manner.

The invention also has the object of providing a process for the preparation of these materials.

A further object of the invention is that of providing electrical conductors which incorporate the aforementioned materials.

Other objects and advantages of the invention will become apparent in the description of the invention which will be exposed hereunder.

DESCRIPTION OF THE INVENTION

The present invention refers to a material useful as an electrical conductor which comprises a conducting polymer incorporated in channels within a porous host, in which the polymer is polyacetylene or a polyacetylene derivative, in which the polymer is formed by polymerization of vapours of

the acetylene monomer, previously adsorbed in channels free of host structure directing agent, with the host encapsulating the polymer in such a way as to protect the polymer from oxidation by ambient oxygen, and where the host is a modified mesoporous aluminosilicate which presents a mean pore diameter between 2 nm and 100 nm.

In a preferred embodiment of the invention, the host is an aluminosilicate with a mean pore diameter of 3.2 nm. In another preferred embodiment, the mean pore diameter of the host is 6.2 nm.

Specially suitable hosts are those constituted by the MCM-41 type hexagonal-phase aluminosilicates or the MCM-48 type cubic-phase ones.

In a preferred embodiment of the invention the aluminosilicate is modified by ion-exchange with transition metal ions, or by impregnation by the introduction of transition metal ions. Such metallic ions can be Fe^{3+}, Cu^{2+} and, especially Ni^{2+} ions. $Fe(NO_3)_3$, $Cu(AcO)_2$ and $Ni(AcO)_2$ are particularly suitable sources of these ions.

According to what has been stated above, the conducting organic polymers are polyacetylene and polyacetylenes substituted with, for example, methyl, phenyl, para-substituted phenyls, cyano and methoxycarbonyl. Said polymers can be present in the neutral state and become activated, for the corresponding use, by adding positive charges consisting in a p-doping or negative charges consisting in an n-doping.

The invention also refers to a process for preparing the aforementioned materials, which consists in incorporating a conducting polymer into channels within a porous host, in which the polymer is polyacetylene or a polyacetylene derivative, which process is described below.

In a first stage, a host is selected constituted by a modified mesoporous aluminosilicate, which presents a mean pore diameter between 2 nm and 100 nm. According to what was stated above in relation to the description of the invention, aluminosilicates of the MCM-41 and MCM-48 types can be employed. These aluminosilicates can be obtained by means of

conventional methods basically consisting in a hydrothermal crystallisation of a mixture of silicate and aluminate gels in the presence of concentrated alkyltrimethylammonium type surfactant solutions (Beck, Vartuli, et al.: "A New Family of Mesoporous Molecular Sieves Prepared with Liquid Crystal Templates" (1992). J. Am. Chem. Soc., 114: 10834-10843; Behrens: "Mesoporous Inorganic Solids" (1993), Adv. Mater. 5: 127-132; Behrens and Haak: "Ordered Molecular Arrays as Templates: A New Approach to Synthesis of Mesoporous Materials" (1993), Angew. Che, Int. Ed. Engl., 32(5): 696-699). In these methods the length of the chain of the alkyl group of alkyltrimethylammonium, concentration of the surfactant and the crystallisation time exert an influence upon the pore size of the final aluminosilicate. In a typical embodiment of these processes, a mixture of ketyltrimethylammonium bromide and tetramethylammonium hydroxide (approximately 15% by weight) is used, which is maintained, without stirring, in an autoclave with a steel reactor coated with TEFLON at approximately 130°C, at autogenous pressure for 24 hours. Higher concentrations of the surfactant agent or longer crystallisation times usually lead to expanded mesoporous materials with a greater mean pore diameter. In order to obtain aluminosilicates useful in relation to the present invention, the parameters of the method for preparing mesoporous aluminosilicates are chosen so that a suitable pore size is obtained, for example, between 2.5 and 6.2 nm, with Si/Al ratios of between ∞ and 13 for example.

To carry out the process of the invention, the structure directing agent usually present in the channels of the aluminosilicate must have been eliminated form the aluminosilicate. The elimination of the structure directing agent can be carried out by means of exhaustive extraction with a suitable solvent. Thus, when the structure directing agent, as is the case with aluminosilicates of the MCM-41 and MCM-48 types, is constituted by alkyltrimethylammonium cations, a suitable solvent is methanol. During the

extraction, the degree of progress of the extraction is verified by combustion analysis (C and N). Alternately, the elimination of the structure directing agent present in the aluminosilicate can be carried out by means of controlled calcination in an oven using gentle temperature gradients up to 550°C, first under a nitrogen current and, in the final phase, under an air current. Both in the case of elimination by solvent extraction as in that employing controlled calcination, the operation is considered finished when the C content, measured by combustion analysis, is below 0.05% by weight compared with the initial value. Typically, aluminosilicates of the MCM-41 and MCM-48 types have an initial organic matter content of approximately 54% by weight.

It must be taken into account that, as is well known, in a large number of mesoporous aluminosilicates, the elimination of the structure directing agent brings about the progressive collapse of the mesopores which can result in a complete collapse of the structure within a matter of months. Therefore, in mesoporous aluminosilicates of this type, it is convenient to use them as hosts in the manufacture of the materials of the present application before approximately 15 days have elapsed after the elimination of the structure directing agent.

After being stripped of the structure directing agent, in a first phase of the process, the aluminosilicate is subjected to dehydration. This dehydration can be carried out by heating the aluminosilicate to approximately 300°C at a vacuum pressure of approximately 10^{-2} torr for 24 hours.

In a second stage, a gaseous current of an acetylene containing monomer is prepared, such as for example acetylene or a substituted acetylene, conveniently treated to ensure it is free of impurities, moisture or other residual gases.

In a third stage, a treatment is carried out consisting in the impregnation of the aluminosilicate with the monomer vapours, so that the aluminosilicate channels become filled with the monomers. Once these have become adsorbed in the

aluminosilicate channels, the monomers polymerise. Conveniently, in order to obtain more uniform materials, the aluminosilicate is magnetically stirred during impregnation with the vapours of the polymer. Although polymerization takes place to a certain extent, at room temperature, it is convenient to carry out this third stage at a temperature between 25 and 400°C in order to achieve maximal incorporation and polymerization of the polymers. Typically, this treatment is carried out in the vapour phase at ambient conditions or at a high vapour pressure, in a reactor, such as for instance a Schlenk type reactor, which contains the aluminosilicate. After finishing the treatment the reactor is emptied under a vacuum. This treatment can be repeated until the necessary degree of impregnation has been achieved, or until maximum impregnation has been achieved. In practice, maximum impregnation has been achieved by repeating the third stage for a total of three times. At the end of this third stage, the material comprised by the aluminosilicate and the polymer is allowed to cool down to room temperature.

The reaction time has an influence upon the achieved polymerization degree. Thus, as the reaction proceeds, a change can be observed in the mesoporous aluminosilicate from a white initial colouring towards a grey or to a black colour. Taking into consideration that polyacetylene is essentially black, the darkening degree of the impregnated host during the reaction is indicative of the polymerisation degree of acetylene into polyacetylene.

In the case of commercial non-substituted acetylene, in order to eliminate acetone, moisture and other impurities that might be present, it is convenient to bubble the gas through a scrubber containing concentrated sulphuric acid, and subsequently through NaOH.

According to what has been indicated above, the aluminosilicate is preferably modified by ion exchange with transition metal ions in an aqueous solution or by impregnation by the introduction of transition metal ions.

The modification by ion-exchange is preferably carried

out when the aluminosilicate contains Al in high amounts and it basically consists in a treatment in which the aluminosilicate is contacted with a concentrated aqueous solution which contains some of the transition metals specified above. In a preferred embodiment of the invention, this solution is of $Ni(AcO)_2$ and typically at a concentration between 0.4-0.8 M, employing, for example, a solid/liquid ratio of approximately 1:10. The contact time between the aluminosilicate and an aqueous solution of the type described above should be conveniently below 30 minutes in order to reduce to a minimum the negative influence of the solution upon the aluminosilicate structure. When higher exchange levels are desired, it is possible to again put the aluminosilicate into contact with a more concentrated solution, typically 1M. This ion exchange treatment to modify the aluminosilicate can also be analogously applied when a transition metal ion different from Ni is chosen.

On the other hand, when dealing with an aluminosilicate in which the Si/Al ratio is ∞, its modification is preferably carried out by an impregnation treatment in which the aluminosilicate and the compound source (for example $Fe(NO_3)_3$, $Cu(AcO)_2$ or, preferably $Ni(AcO)_2$) which contains the transition metal ion, in a ratio between 100:5 and 100:0.1, are mechanically mixed, progressively heating the resulting mixture, preferably up to 300°C.

It has been possible to demonstrate that the formation of the polymers in the aluminosilicate mesoporous host is especially effective when the aluminosilicate exhibits a high exchange degree.

The final structure of the aluminosilicate exchanged with transition metal ions is characterised by a specific x-ray pattern indicative of its crystalline nature and by a certain isothermal gas adsorption capacity indicative of the mesoporous nature and of its surface area. It can usually be observed that the modification of aluminosilicates modified by ion exchange with transition metal ions, gives rise to a reduction in the crystallinity values of up to over 40% and

in surface area of up to over 10% with respect to the values corresponding to non-modified aluminosilicates. By way of example, the MCM-41 material with a pore size of 3.2 nm undergoes, after two successive exchanges with $Ni(AcO)_2$, a 50% reduction in the number of 2θ 2.4° peak counts and a reduction in surface area (according to the BET algorithm) from an initial value of 850 m^2/g to 700 m^2/g. Even so, they still exhibit a high mesoporosity level and are ideal as hosts in the materials of the present invention.

The rigid matrix of the aluminosilicate employed according to the invention contains active centres that act as catalytic centres of the polymerization process. After polymerization, the aluminosilicate channels are packed with the polymer chains and its inert inorganic walls act as protective sheaths. In this way, the tendency of the polymers towards decomposing is drastically reduced. This implies that these materials can be stored and processed in environments in which oxygen is present without any loss in their properties.

The doping of the conductive materials of the present invention, that is, the oxidation or reduction of the polymers encapsulated within the mesoporous aluminosilicates, consists in charging the encapsulated polymer positively by means of a p-type doping or negatively by means of n-type doping. Doping can be carried out by chemical or by electrochemical means.

For the electrochemical doping of the materials of the present invention, modified electrodes are prepared by conventional methods, finally applying a positive electrical potential (for p-doping) or a negative electrical potential (for n-doping).

To obtain a chemically doped material of the p-type, the starting material consists of mesoporous aluminosilicates exchanged with Cu^{2+} since, after polymerization, Cu^{2+} becomes reduced to Cu^{+}, which in turn implies the oxidation of the polymer. Sn^{4+} or other metallic ions that can undergo reduction after the formation of the polymer can also be

used.

On the other hand, in order to obtain a doped material of the n-type, the starting materials are mesoporous aluminosilicates which contain Ni^{2+} and which are subjected to a prior reduction under hydrogen current at approximately 500°C. By means of this reduction, the transformation of Ni^{2+} into Ni(0) is achieved, being it possible to visually appreciate this transformation by the progressive darkening of the aluminosilicate which at the end becomes completely black. The aluminosilicate containing Ni(0) atoms must be stored and handled in an inert atmosphere in the absolute absence of moisture or oxygen. The polymerization of the monomers in Ni(0) based mesoporous aluminosilicate hosts is carried out following the process described above.

The invention also refers to electrical conductors which comprise the materials described above. These conductors can be, for example, electrodes for batteries, especially lithium batteries, conducting surfaces for displays, conducting surfaces for luminous devices, and the like.

Typically, an electrode according to the invention is prepared by coating a conductive, inert, vitrified and polished host, with a paste composed at least by a mixture of the conducting material, powdered graphite and paraffin with an agglutinating agent, for example in a ratio by weight between 80:15:5 and 10:85:5, and allowing the electrode so obtained to dry. In general, lower ratios of the polymer-containing aluminosilicate and greater ones of graphite, such as for example 50:40:10 remarkably decrease the resistivity of the paste. A preferred preparation of the invention employs a ratio between polymer-containing aluminosilicate: graphite:paraffin of 35:60:5.

The conducting surfaces according to the invention are typically obtained from the deposition, on an inert conducting host, of a layer of a suspension of the polyacetylene-containing mesoporous host according to the invention in an organic solvent, such as for example acetone, adding, after allowing to air-dry, an aqueous solution, for

example at 10%, of polyvinyl alcohol.

Conductors so prepared, be them in paste or layer form, are subjected to a positive electrical potential (for example +2,0 V, for p-doping) or to a negative electrical potential (for example - 1,7 V, for n-doping), to respectively achieve the polymer oxidation or reduction.

The mesoporous aluminosilicates which are used in the preparation of the conductive materials of the present application, have the advantage of having an internal volume and an internal surface far greater to that of the zeolites and, therefore, permit the effective encapsulation of amounts greater than 30% of polyacetylene and polyacetylene derivatives, sufficient for a great part of industrial applications.

DETAILED DESCRIPTION OF THE INVENTION

The following examples will illustrate the invention in greater detail.

Example 1:

The alkyltrimethylammonium ion content present as structure directing agent, was determined by solids combustion analysis of ten samples of mesoporous aluminosilicates of the MCM-41 type, with pore sizes of 3.2 nm and 6.2 nm respectively, and mean particle sizes of 1 μm. A content of approximately 55% by weight was detected.

The samples were subjected to exhaustive conventional extraction, using methanol as a solvent to eliminate the structure directing agent, checking the progress of the elimination by C combustion analysis.

Subsequently the Ni^{2+} ions were introduced by successively contacting the solids with two 0.6 and 1.0 M aqueous solutions of $Ni(AcO)_2$ for 20 minute periods. The final content of Ni^{2+} was around 3% by weight for the 3.2 nm aluminosilicate and 0.5 for the 6.2 nm aluminosilicate.

Subsequently the samples were subjected to dehydration by heating at 300°C at a pressure of 10^{-2} torr for 24 hours.

Example 2:

Five samples of MCM-41 with a mean pore size of 3.2 nm

and another five samples of MCM-41 with a mean pore size of 6.2 nm were placed into independent vials of 5 ml of capacity which are hermetically attached to a Schlenk type reactor, provided with a drum which permits the simultaneous treatment of five samples at controlled temperature, into which commercial acetylene gas was introduced, which had previously been purified by bubbling through a container with concentrated sulphuric acid and subsequently through a second container with NaOH, at a pressure of 1 atm and for a period of five minutes, and were subjected to different temperatures. Finally the reactor was effluxed to vacuum and the samples were allowed to cool at room temperature. This cycle was repeated three times.

It was possible to observe that temperature exerted a crucial influence upon the polymerization of acetylene in the reactor. Therefore it was possible to quantitatively establish the amount of polymer formed by means of combustion analysis of the solid to determine the total C content of the obtained materials. The results of the analyses are shown in table I.

Table 1: Effect of the reaction temperature upon the carbon content for the reaction of acetylene in the presence of mesoporous aluminosilicates.

	C Content (% by weight)	
Temperature (°C)	MCM-41 (3.2 nm)	MCM-48 (6.2 nm)
Ambient	5.88	3.91
90	6.47	5.94
150	26.39	2.06
225	32.52	27.51
300	51.65	29.62

According to what is shown by the values indicated in table I, the maximum content achieved for polyacetylene if 51%. This value is remarkable and allows applications such as the use of the material as electrodes in lithium batteries

which demand a high content in the electroactive component. In fact, when the C content in the initial samples (54% by weight of alkyltrimethylammonium) is compared with the value for 3.2 nm MCM-41 at a temperature of 300°C, it is possible to observe that most part of the mesoporous aluminosilicate channels, which were initially occupied by the structure directing agent, have been occupied by polyacetylene.

According to the current knowledge of mesoporous materials, the maximum packing density of an organic host within the channels, corresponds to that of the structure directing agent. Accordingly, it is possible to conclude that by means of the process of the present invention, an almost complete filling of the internal spaces of the aluminosilicate was achieved, with an efficacy and packaging degree similar to that of the structure directing agent.

Example 3:

The samples prepared according to example 2 were analysed with regard to the presence and purity of the polyacetylene synthesised within the interior of the mesoporous aluminosilicate. It was possible to establish that

i) the FT-Raman spectra of the samples coincide with those described in the chemical literature (Lewis, Millar et al.: "Trans-Polyacetylene on Sodium and Cesium Mordenites: A Resonance Raman Spectroscopic Study" (1993), Chem. Mat., 5:1509-1517; Millar, Lewis et al: "Raman-Spectroscopic Study of the Formation of Polyacetylene within Zeolite Channels" (1993), J. Mater. Chem., 3:867-872) for polyacetylenes, being the most characteristic absorptions in the bands of 1490 and 1110 cm^{-1};

ii) the UV/vis spectra obtained by diffuse reflectance coincide with those described (Cox and Stucky: "Polymerization of Methylacetylene in Hydrogen Zeolites" (1991), J. Phys. Chem., 95:710-720; Pereira, Kokotailo et al.: "Acetylene Polymerization in a H-ZSM-5 Zeolite" (1991), J. Mater. Chem., 3(8):867-872) for the polyacetylenes and are characterised by absorption throughout the whole range of wavelengths, in which the extinction coefficient decreases

progressively with the increase in wavelength;

iii) in solids ^{13}C NMR spectroscopy signals appear which correspond to olefinic carbons between 150 and 100 ppm, together with the signals characteristic of each constituent if any;

iv) in X-ray photoelectronic spectroscopy (XPS) it is possible to confirm the presence, in the external surface, of carbon aluminosilicate particles with sp^2 hybridisation, which is indicative of the presence of polyacetylene;

v) in mass spectroscopy using the fast atom bombardment method (FAB-MS) it is established that the maximum weight of the polymer situated in the outermost parts of the aluminosilicate particle is greater than 1200 dalton;

vi) when the polymer content of the samples was established by thermogravimetry coupled with differential scanning calorimetry it was established that the encapsulated polyacetylenes are thermally stable up to a temperature of 300°C.

Example 4:

In order to obtain an electrode comprising 6.2 nm MCM-41 obtained according to example 2, for a lithium battery, the following process is carried out:

80g of aluminosilicate are mixed in a mortar with 15g of graphite powder and 5g of paraffin are added as agglutinant (commercialised by Aldrich under the name Nujol oil for spectroscopy) to obtain a paste which is deposited on a conveniently polished inert conducting host, and allowed to dry.

The so formed electrode is subjected to +2.0V for a p-doping, or alternatively, to -1.7V for an n-doping, to achieve its complete oxidation or reduction, respectively. In the electrochemical process Pt is employed as ancillary electrode, and a saturated solution of calomel as reference electrode. Although the doping can be carried out in an aqueous medium, it can also be carried out in organic solvents such as acetonitrile and dimethylformamide. Among the supporting electrolytes that can be used in organic

solvents it is possible to mention tetraethylammonium perchlorate, n-butylammonium hexafluorophosphate, whereas lithium or sodium perchlorate can be used in water.

Example 5:

In order to obtain a conducting surface comprising 6.2 nm MCM-41 obtained according to example 2, a suspension of 1g of MCM-41 is prepared in 25 ml of acetone and is deposited over an inert conductive host. After air-drying, few drops of an aqueous solution of 10% polyvinyl alcohol are added.

The so formed electrode is subjected to +2.0V for a p-doping, or alternatively, to -1.7V for an n-doping, to achieve its complete oxidation or reduction, respectively. The electrochemical process is carried out using the same means and reagents as those described for example 3.

CLAIMS

1. A material useful as an electrical conductor comprising a conducting polymer incorporated in channels within a porous host, wherein the polymer is polyacetylene or a polyacetylene derivative,

characterised in that

the polymer is formed by polymerization of vapours of the acetylene monomer, previously adsorbed in channels of the mesoporous material, free of host structure directing agent;

the host encapsulates the polymer in such a way that the polymer is protected against oxidation by ambient oxygen; and

the host is a modified mesoporous aluminosilicate which presents a mean pore diameter between 2 nm and 100 nm.

2. A host according to claim 1, characterised in that the host is an aluminosilicate with a mean pore diameter of 3.2 nm.

3. A host according to claim 1, characterised in that the host is an aluminosilicate with a mean pore diameter of 6.2 nm.

4. A material according to claims 1, 2 or 3, characterised in that it comprises an aluminosilicate host which incorporates a polymer comprised by polyacetylene substituted with phenyl groups.

5. A material according to any of claims 1 to 4, characterised in that the host is an hexagonal-phase aluminosilicate of the MCM-41 type.

6. A material according to any of claims 1 to 4, characterised in that the host is a cubic-phase aluminosilicate of the MCM-48 type.

7. A material according to any of claims 1 to 6, characterised in that the aluminosilicate has been modified by ion-exchange with transition metal ions.

8. A material according to any of claims 1 to 6, characterised in that the aluminosilicate has been modified by impregnation by means of the introduction of transition metal ions.

9. A material according to claims 7 or 8, characterised in that the metal ions are Ni^{2+} ions.

10. A material according to claim 9, characterised in that the transition metal ions originate from $Ni(AcO)_2$.

11. A material according to claims 7 or 8, characterised in that the metal ions are Fe^{3+} ions.

12. A material according to claim 11, characterised in that the Fe^{3+} ions originate from $Fe(NO_3)_3$.

13. A material according to claims 7 or 8, characterised in that the metal ions are Cu^{2+} ions.

14. A material according to claim 13, characterised in that the transition metal ions originate from $Cu(AcO)_2$.

15. A material according to claims 1 to 4, characterised in that the polymer exhibits a positive charge consisting in a p-doping.

16. A material according to claims 1 to 4, characterised in that the polymer exhibits a negative charge consisting in an n-doping.

17. A process to prepare an electrically conductive material which comprises

incorporating a conductive polymer into channels within a porous host, in which the polymer is polyacetylene or a polyacetylene derivative,

characterised in that it comprises

dehydrating a modified aluminosilicate not containing a structure directing agent in its channels;

preparing a vapour of an acetylene monomer;

performing at least one impregnation and polymerization phase which comprises incorporating the vapour into the aluminosilicate channels, and subjecting the aluminosilicate with the monomers to polymerization by means of a thermal treatment at a temperature between 25 and 400°C;

the host being a modified mesoporous aluminosilicate which presents a mean pore diameter between 2 nm and 100 nm.

18. A process according to claim 17, characterised in that the host is selected from a MCM-41 type hexagonal-phase aluminosilicate and a MCM-48 type cubic-phase

aluminosilicate.

19. A process according to any of claims 16 to 18, characterised in that the aluminosilicate is modified by ion-exchange with transition metal ions in an aqueous solution prior to the polymerization phase.

20. A process according to any of claims 16 to 18, characterised in that the aluminosilicate is modified by impregnation by means of the introduction of transition metal ions.

21. A process according to claim 19 or 20, characterised in that the metal ions are selected from Ni^{2+}, Fe^{3+} and Cu^{2+} ions.

22. A process according to claim 21, characterised in that the Ni^{2+} ions originate from $Ni(AcO)_2$.

23. A process according to claim 21, characterised in that the Fe^{2+} ions originate from $Fe(NO_3)_3$.

24. A process according to claim 21, characterised in that the Cu^{2+} ions originate from $Cu(AcO)_2$.

25. A process according to claim 17, characterised in that the encapsulated polymer is positively charged by means of a p-type doping.

26. A process according to claim 17, characterised in that the encapsulated polymer is negatively charged by means of a n-type doping.

27. A process according to claims 25 or 26, characterised in that the doping is carried out electrochemically.

28. A process according to claims 25 or 26, characterised in that the doping is carried out chemically.

29. A process according to claim 17, characterised in that the impregnation and polymerization phase is repeated three times.

30. A process according to claim 17, characterised in that the polymer is polyacetylene substituted with methyl, phenyl, para-substituted phenyls, cyano, methoxycarbonyl groups.

31. An electrical conductor characterised in that it

comprises a material according to any of claims 1-16.

32. An electrical conductor characterised in that it comprises a material prepared according to the process according to any of claims 17-30.

33. A conductor according to claims 31 or 32, characterised in that it is an electrode used in batteries, which comprises said material.

34. A conductor according to claims 31 or 32, characterised in that it is a conductive surface for displays, which comprises said material.

35. A conductor according to claims 31 or 32, characterised in that it is a conductive surface for luminous devices, which comprises said material.

36. A conductor according to claim 33, characterised in that it is an electrode in lithium batteries, which comprises said material.

37. A conductor according to claims 33 or 36, characterised in that the electrode comprises

an inert, vitrified and polished electrode

a coating of the inert electrode at least composed by a mixture of said material, powdered graphite and paraffin as an agglutinant.

38. A conductor according to claim 37, characterised in that the coating contains said material, graphite and paraffin in a ratio by weight of 80:15:5.

39. A conductor according to claims 34 or 35, characterised in that the conducting surface is a layer obtained by the deposition on a host of a suspension of said material on an organic solvent with an aqueous solution of polyvinyl alcohol.

40. A conductor according to claim 39, characterised in that the organic solvent is acetone.

41. A conductor according to claim 39, characterised in that the aqueous solution of polyvinyl alcohol is a 10% solution.

ABSTRACT

A material useful as an electrical conductor which comprises a conducting polymer incorporated in channels within a porous host, in which the polymer is polyacetylene or a polyacetylene derivative, in which the polymer is formed by polymerization of vapours of monomer acetylene, previously adsorbed in channels free of structure directing agent of the host, which provides an encapsulation to the polymer which protects the polymer against oxidation by oxygen in the environment, being the host a modified mesoporous aluminosilicate which presents a mean pore diameter between 2 nm and 100 nm, such as a MCM-41 type hexagonal-phase aluminosilicate or a MCM-48 type cubic-phase aluminosilicate.

A process is also described for the preparation of this material and conducting materials, for example electrodes for batteries and conducting surfaces, which comprise these materials.

1007057.070902 ① #6

Practitioner's Docket No. U 013829-3

PATENT

Optional Customer No. Bar Code

00140

PATENT TRADEMARK OFFICE

COMBINED DECLARATION AND POWER OF ATTORNEY

(ORIGINAL, DESIGN, NATIONAL STAGE OF PCT, SUPPLEMENTAL, DIVISIONAL, CONTINUATION, OR C-I-P)

As a below named inventor, I hereby declare that:

TYPE OF DECLARATION

This declaration is of the following type:

(check one applicable item below)

[ ] original.
[ ] design.

NOTE *With the exception of a supplemental oath or declaration submitted in a reissue, a supplemental oath or declaration is not treated as an amendment under 37 CFR 1 312 (Amendments after allowance). M.P.E.P Section 714.16, 7th Ed.*

[ ] supplemental.

NOTE *If the declaration is for an International Application being filed as a divisional, continuation or continuation-in-part application, do not check next item; check appropriate one of last three items.*

[X] national stage of PCT.

NOTE: *If one of the following 3 items apply, then complete and also attach ADDED PAGES FOR DIVISIONAL, CONTINUATION OR C-I-P.*

NOTE: *See 37 C F R. Section 1.63(d) (continued prosecution application) for use of a prior nonprovisional application declaration in the continuation or divisional application being filed on behalf of the same or fewer of the inventors named in the prior application.*

[ ] divisional.
[ ] continuation.

NOTE *Where an application discloses and claims subject matter not disclosed in the prior application, or a continuation or divisional application names an inventor not named in the prior application, a continuation-in-part application must be filed under 37 C.F.R. Section 1.53(b) (application filing requirements-nonprovisional application)*

[ ] continuation-in-part (C-I-P).

EXPRESS MAIL LABEL NO.: EV 011023081 US

(check proper box(es) for any of the following added page(s) that form a part of this declaration)

[X] **Signature** for fourth and subsequent joint inventors. *Number of pages added* ____1____

* * *

[ ] **Signature** by administrator(trix), executor(trix) or legal representative for deceased or incapacitated inventor. *Number of pages added* _____

* * *

[ ] **Signature** for inventor who refuses to sign or cannot be reached by person authorized under 37 C.F.R. Section 1.47. *Number of pages added* _____

* * *

[ ] Added page for **signature** by one joint inventor on behalf of deceased inventor(s) where legal representative cannot be appointed in time. (37 C.F.R. Section 1.47)

* * *

[ ] Added pages to combined declaration and power of attorney for divisional, continuation, or continuation-in-part (C-I-P) application.

[ ] Number of pages added _____

* * *

[ ] Authorization of practitioner(s) to accept and follow instructions from representative.

(If no further pages form a part of this Declaration, then end this Declaration with this page and check the following item)

[ ] This declaration ends with this page.

INVENTORSHIP IDENTIFICATION

WARNING: *If the inventors are each not the inventors of all the claims, an explanation of the facts, including the ownership of all the claims at the time the last claimed invention was made, should be submitted*

My residence, post office address and citizenship are as stated below, next to my name. I believe that I am the original, first and sole inventor *(if only one name is listed below)* or an original, first and joint inventor *(if plural names are listed below)* of the subject matter that is claimed, and for which a patent is sought on the invention entitled:

TITLE OF INVENTION

MATERIAL USEFUL AS ELECTRIC CONDUCTOR AND COMPRISING POLYACETYLENE OR SUBSTITUTED POLYACETYLENES INCORPORATED IN A MESOPOROUS SUPPORT, PRODUCTION PROCESS, AND CONDUCTORS MADE OF SAID MATERIAL

SPECIFICATION IDENTIFICATION

The specification of which:

(complete (a), (b), or (c))

(a) [ ] is attached hereto.

NOTE "The following combinations of information supplied in an oath or declaration filed on the application filing date with a specification are acceptable as minimums for identifying a specification and compliance with any one of the items below will be accepted as complying with the identification requirement of 37 C.F R Section 1 63:

"(1) name of inventor(s), and reference to an attached specification which is both attached to the oath or declaration at the time of execution and submitted with the oath or declaration on filing,

"(2) name of inventor(s), and attorney docket number which was on the specification as filed; or

"(3) name of inventor(s), and title which was on the specification as filed "

Notice of July 13, 1995 (1177 O G. 60)

(b) [ ] was filed on _____, [ ] as Application No. _____
[ ] and was amended on _____ (if applicable).

NOTE· Amendments filed after the original papers are deposited with the PTO that contain new matter are not accorded a filing date by being referred to in the declaration. Accordingly, the amendments involved are those filed with the application papers or, in the case of a supplemental declaration, are those amendments claiming matter not encompassed in the original statement of invention or claims See 37 C F.R. Section 1 67.

NOTE. "The following combinations of information supplied in an oath or declaration filed after the filing date are acceptable as minimums for identifying a specification and compliance with any one of the items below will be accepted as complying with the identification requirement of 37 C.F.R Section 1 63

(A) application number (consisting of the series code and the serial number, e g., 08/123,456),
(B) serial number and filing date,
(C) attorney docket number which was on the specification as filed,
(D) title which was on the specification as filed and reference to an attached specification which is both attached to the oath or declaration at the time of execution and submitted with the oath or declaration; or
(E) title which was on the specification as filed and accompanied by a cover letter accurately identifying the application for which it was intended by either the application number (consisting of the series code and the serial number, e.g., 08/123,456), or serial number and filing date Absent any statement(s) to the contrary, it will be presumed that the application filed in the PTO is the application which the inventor(s) executed by signing the oath or declaration

M.P.E.P. § 601.01(a), 7th ed.

(c) [X] was described and claimed in PCT International Application No. PCT/ES00/00260 filed on 19 JULY 2000 and as amended under PCT Article 19 on _____ *(if any).*

SUPPLEMENTAL DECLARATION (37 C.F.R. Section 1.67(b))

(complete the following where a supplemental declaration is being submitted)

[ ] I hereby declare that the subject matter of the

[ ] attached amendment
[ ] amendment filed on _____.

was part of my/our invention and was invented before the filing date of the original application, above identified, for such invention.

ACKNOWLEDGMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information, which is material to patentability as defined in 37, Code of Federal Regulations, Section 1.56,

(also check the following items, if desired)

[ ] and which is material to the examination of this application, namely, information where there is a substantial likelihood that a reasonable Examiner would consider it important in deciding whether to allow the application to issue as a patent, and

[ ] in compliance with this duty, there is attached an information disclosure statement, in accordance with 37 C.F.R. Section 1.98.

PRIORITY CLAIM (35 U.S.C. Section 119(a)-(d))

NOTE: 37 C F.R § 1 55 Claim for foreign priority.

"(a) An applicant in a nonprovisonal application may claim the benefit of the filing date of one or more prior foreign applications under the conditions specified in 35 U.S C 119(a) through (d) and (f), 172, and 365(a) and (b).

(1)(i) In an original application filed under 35 U S C 111(a), the claim for priority must be presented during the pendency of the application,, and within the later of four months from the actual filing date of the application or sixteen months from the fling date of the prior foreign application. This time period is not extendable. The claim must identify the foreign application for which priority os claimed, as well as any foreign application for the same subject matter and having a filing date before that of the application for which priority is claimed, by specifying the application number, country (or intellectual property authority), day, month, and year of its filing The time period in this paragraph does not apply to an application for a design patent.

(ii) In an application that entered the national stage from an international application after compliance with 35 U.S.C. 371, the claim for priority must be made during the pendency of the application and within the time limit set forth in the PCT and the Regulations under the PCT."

(2) The claim for priority and the certified copy of the foreign application specified in 35 U S C. 119(b) or PCT Rule 17 must, in any event, be filed before the patent is granted If the claim for priority or the certified copy of the foreign application is filed after the date the issue fee is paid, it must be accompanied by the processing fee set forth in § 1.17(i), but the patent will not include the priority claim unless corrected by a certificate of correction under 35 U S C 255 and § 1.323.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

(complete (d) or (e))

(d) [ ] no such applications have been filed.
(e) [X] such applications have been filed as follows.

NOTE: Where item (c) is entered above and the International Application which designated the U S. itself claimed priority check item (e), enter the details below and make the priority claim

PRIOR FOREIGN/PCT APPLICATION(S) FILED WITHIN 12 MONTHS (6 MONTHS FOR DESIGN) PRIOR TO THIS APPLICATION AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. SECTION 119(a)-(d)

COUNTRY (OR INDICATE IF PCT)	APPLICATION NUMBER	DATE OF FILING DAY, **MONTH**, YEAR	PRIORITY CLAIMED UNDER 35 USC 119
ES	P 9901708	21 JULY 1999	[X]YES [ ]NO
			[ ]YES [ ]NO
			[ ]YES [ ]NO
			[ ]YES [ ]NO
			[ ]YES [ ]NO

CLAIM FOR BENEFIT OF PRIOR U.S. PROVISIONAL APPLICATION(S)
(35 U.S.C. Section 119(e))

I hereby claim the benefit under Title 35, United States Code, Section 119(e) of any United States provisional application(s) listed below:

PROVISIONAL APPLICATION NUMBER **FILING DATE**

_____ / _____ _____
_____ / _____ _____
_____ / _____ _____

CLAIM FOR BENEFIT OF EARLIER U.S./PCT APPLICATION(S) UNDER 35 U.S.C. SECTION 120

[ ] The claim for the benefit of any such applications are set forth in the attached ADDED PAGES TO COMBINED DECLARATION AND POWER OF ATTORNEY FOR DIVISIONAL, CONTINUATION OR CONTINUATION-IN-PART (C-I-P) APPLICATION.

ALL FOREIGN APPLICATION(S), *IF ANY*, FILED MORE THAN 12 MONTHS (6 MONTHS FOR DESIGN) PRIOR TO THIS U.S. APPLICATION

_____

_____

_____

NOTE *If the application filed more than 12 months from the filing date of this application is a PCT filing forming the basis for this application entering the United States as (1) the national stage, or (2) a continuation, divisional, or continuation-in-part, then also complete ADDED PAGES TO COMBINED DECLARATION AND POWER OF ATTORNEY FOR DIVISIONAL, CONTINUATION OR C-I-P APPLICATION for benefit of the prior U.S. or PCT application(s) under 35 U.S.C. Section 120.*

POWER OF ATTORNEY

I hereby appoint the following practitioner(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

(list name and registration number)

(10)

JOSEPH H. HANDELMAN, 26179

JOHN RICHARDS, 31053

RICHARD J. STREIT, 25765

PETER D. GALLOWAY, 27885

RICHARD P. BERG, 28145

JULIAN H. COHEN, 20302

WILLIAM R. EVANS 25858

JANET I. CORD, 33778

CLIFFORD J. MASS, 30086

CYNTHIA R. MILLER, 34678

(Check the following item, if applicable)

[ ] I hereby appoint the practitioner(s) associated with the Customer Number provided below to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

[ ] Attached, as part of this declaration and power of attorney, is the authorization of the above-named practitioner(s) to accept and follow instructions from my representative(s).

NOTE: "Special care should be taken in continuation or divisional applications to ensure that any change of correspondence address in a prior application is reflected in the continuation or divisional application For example, where a copy of the oath or declaration from the prior application is submitted for a continuation or divisional application filed under 37 CFR 1.53(b) and the copy of the oath or declaration from the prior application designates an old correspondence address, the Office may not recognize, in the continuation or divisional application, the change of correspondence address made during the prosecution of the prior application Applicant is required to identify the change of correspondence address in the continuation or divisional application to ensure that communications from the Office are mailed to the current correspondence address 37 CFR 1 63(d)(4) " Section 601.03, M P E P., 7th Ed

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SEND CORRESPONDENCE TO

Ladas & Parry
26 West 61st Street
New York, N.Y. 10023

DIRECT TELEPHONE CALLS TO:
(Name and telephone number)

John Richards
(212) 708-1915

(complete the following if applicable)

Since this filing is a [ ] continuation [ ] divisional there is attached hereto a Change of Correspondence Address so that there will be no question as to where the PTO should direct all correspondence.

DECLARATION

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

SIGNATURE(S)

NOTE: Carefully indicate the family (or last) name, as it should appear on the filing receipt and all other document.

NOTE: Each inventor must be identified by full name, including the family name, and at least one given name without abbreviation together with any other given name or initial, and by his/her residence, post office address and country of citizenship. 37 C.F.R. Section 1.63(a)(3)

NOTE: Inventors may execute separate declarations/oaths provided each declaration/oath sets forth all the inventors. Section 1.63(a)(3) requires that a declaration/oath, inter alia, identify each inventor and prohibits the execution of separate declarations/oaths which each sets forth only the name of the executing inventor. 62 Fed. Reg. 53,131, 53,142, October 10, 1997.

1-00

Full name of sole or first inventor

Mercedes / *(Given Name)* ____ / *(Middle Initial or Name)* ALVARO RODRIGUEZ / *Family (Or Last Name)*

Inventor's signature (X) Mercedes Alvaro Rodriguez

Date (X) 26-06-02 **Country of Citizenship** SPAIN

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Camino de Vera, E-46022 Valencia, Spain

2-00

Full name of second joint inventor, if any

Hermenegildo / *(Given Name)* ____ / *(Middle Initial or Name)* GARCIA GOMEZ / *Family (Or Last Name)*

Inventor's signature (X) HERMENEGILDO GARCIA GOMEZ

Date (X) 28-06-02 **Country of Citizenship** SPAIN

Residence Valencia, SPAIN ESX

Post Office Address Universidad Politecnica de Valencia, Departamento de Quimica,
Camino de Vera, E-46022 Valencia, Spain

3-00

Full name of third joint inventor, if any

Andrew / *(Given Name)* ____ / *(Middle Initial or Name)* GILBERT / *Family (Or Last Name)*

Inventor's signature (X) A. McGilbert

Date (X) 19-06-02 **Country of Citizenship** SPAIN

Residence Valencia, SPAIN ESX

Post Office Address Universidad Politecnica de Valencia, Departamento de Quimica,
Camino de Vera, E-46022 Valencia, Spain

Practitioner's Docket No. U 013829-3

ADDED PAGE TO COMBINED DECLARATION AND POWER OF ATTORNEY FOR SIGNATURE BY FOURTH AND SUBSEQUENT INVENTORS

4-00

Full name of fourth joint inventor, if any

David J. CARDIN
(Given Name) *(Middle Initial or Name)* *Family (Or Last Name)*

Inventor's signature (X) David J. Cardin

Date (X) 19-06-02 **Country of Citizenship** SPAIN

Residence Valencia, SPAIN ESX

Post Office Address Universidad Politecnica de Valencia, Departamento de Quimica,
Camino de Vera, E-46022 Valencia, Spain

Full name of fifth joint inventor, if any

(Given Name) *(Middle Initial or Name)* *Family (Or Last Name)*

Inventor's signature _____

Date _____ **Country of Citizenship** _____

Residence _____

Post Office Address _____

Full name of sixth joint inventor, if any

(Given Name) *(Middle Initial or Name)* *Family (Or Last Name)*

Inventor's signature _____

Date _____ **Country of Citizenship** _____

Residence _____

Post Office Address _____